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SEMIANNUAL REPORT

CONTRACT NONR 4849(00)

HIGH POWERED DIFFRACTION LIMITED RAMAN LASER

15 APRIL 1965 through 15 OCTOBER 1965

HUGHES RESEARCH LABORATORIES
Malibu, California

a division of hughes aircraft company

HIGH POWERED DIFFRACTION LIMITED RAMAN LASER

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Quantum Electronics Department

Semiannual Report
15 April through 15 October 1965

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ABSTRACT

The problems associated with the production of diffraction-limited Raman based laser-like devices have been under investigation. The most persistent effort has been directed at understanding several of the rather serious deviations between theory and experiment. The most noteworthy problem treated here is the anomalous gain of Stimulated Raman Scattering (SRS). We have analyzed several approaches, theoretically and experimentally, with some success in only one approach. At present, it appears that the solution is contained in considering light trapping effects as the cause of a highly increased power density in the Raman material. Currently, efforts are continuing to explain the mechanism of the trapping as well as relating these effects to other problems in SRS.

INTRODUCTION

The possibility of producing a diffraction-limited Raman laser cannot be determined at the present time. Laboratory experiments have produced moderately high power lasers with narrow, but not diffraction-limited, beamwidth. With this in mind, we are attempting to solve many of the problems connected with the Raman laser research program. Most of the problems lie in the area of the fundamental understanding of the physical effect. The theory of Stimulated Raman Scattering (SRS) has not yet satisfactorily explained such parameters as the Raman gain, anti-Stokes emission angles, the importance of regeneration, the ratio of forward-to-backward Stokes, or the Stokes-to-anti-Stokes ratio. In addition, the relationships between SRS and such related processes as stimulated Brillouin scattering, optical parametric effects, light trapping, etc., is not clear. Some data have been obtained recently on SRS conversion efficiencies, but other engineering data (such as beam divergences and the possibility of mode control) are lacking.

SUMMARY OF EXPERIMENTAL WORK

Initial work was done in an attempt to clarify the position of the theory of SRS by making a series of measurements of conversion efficiency. From this type of data, rough estimates of the Raman gain can be obtained by the following argument. At threshold, the Raman gain must equal the losses in the system and oscillation will result. The primary source of loss in our experiments was considered reflection loss, since there were no reflectors around the Raman cell. Careful estimates of the diffuse reflectivity showed that a reasonable value for R (the reflection coefficient) would be about 10^{-8}.[1] Now it is necessary that the gain be sufficient to overcome this loss on each double pass of the Raman cell. Typically the cell is 10 cm long (ℓ) and the gain (g_β) can be found by solving $R \exp[g_\beta 2\ell] \geq 1$, or $10^{-8} \exp[20\, g_\beta] \geq 1$. It is then clear that g_β must be of the order of 1 cm^{-1} and this value is very insensitive to the choice of R for a change in R by several orders of magnitude. This value of g_β is to be compared with a theoretical value of 5×10^{-3} cm^{-1} under these conditions. For a more detailed discussion, see Appendix A.

The major discrepancy between theory and experiment has been the subject of intensive study. Several suggestions have been advanced in an attempt to explain the discrepancy. We suggested several solutions[1] one of which was independently analyzed by Bloembergen and Shen.[2] It was suggested that the mode structure of the laser source could cause an increase in the gain of the Raman material by as much as an order of magnitude for most typical lasers. However, when we were able to produce a laser which was essentially single mode, the results were virtually unaltered. See Appendix B. The multimode arguments can be summarized by noting that if the laser consists of many modes, then

there must be minima and maxima in the intensity of the laser, either in time or space. Both arguments yield the same results. The nonlinear process (Raman gain) grows on the peaks in intensity more rapidly than it decays on the minima and the results are as if the wrong intensity were used (the spatial or temporal average) instead of the correct intensity (the local intensity integrated along the path propagating with the growing Raman wave). Although this argument must be correct, it is true that it does not explain the discrepancy between experiment and theory, but rather deepens the puzzle.

Other suggestions have been made to the effect that there is some process producing a multimode character in the laser pump wher there was none before. However, it should be noted that this is not a multimode pump argument, since it is the effect that is discussed, and the cause is still unknown. We suggested several other causes[1] which have not as yet been verified in detail. The first one we will discuss is the least tested concept. The explanation could lie with the stimulated Brillouin scattering. The Brillouin scattering could produce other frequencies from a single frequency laser source and therefore create "temporal modes" where there were none before. Or, the Brillouin scattering may be able to enhance the reflection for the Stokes waves just as it does for the laser and therefore the value of R changes from 10^{-8} to 10^{-1} or thereabouts; then g_β would have to be only slightly larger than the theoretical value. Unfortunately, neither of these possibilities has been tested as yet. Preliminary measurements have failed to show stimulated Brillouin scattering occurring at or before the Raman threshold in the materials that have shown the anomalous gain.

The other suggestion deals with light trapping effects first treated by Chiao, et al.[3] Early efforts to observe such effects were fraught with difficulties. Later efforts have shown some evidence of light trapping effects and have pointed out where the problems occurred in the early measurements. Basically, all that was required was a constriction of the whole beam or sizable regions by factors of 5 or more in linear dimensions. The diffraction spreading of anti-Stokes rings and forward Stokes emission places a lower limit of about 100μ on the size of the constriction that would be expected. Actual observations have shown that this is the right magnitude or, possibly, the constricted region is smaller than 100μ. Under these conditions, the laser pump energy will diffraction-spread out of the constricted region very rapidly after it exits from the Raman cell. In fact, the near field ends less than one cm past the end of the cell and, unfortunately, the early observations were focused in the far field of these constricted "filaments" or guided wave regions. More recent data have shown that the minor changes that were seen earlier were the diffraction-spreading effects in the far field which did not look at all like "hot spots."

It should be pointed out that even though "hot spots" in the laser pump are seen to occur at and around the threshold for SRS, they have

not been positively identified as the cause of the SRS as contrasted with an interpretation that they are a result of SRS. We feel, however, that they are the cause of the SRS. It should be noted that the cause of the trapping apparently observed by us and others[4] has not been identified. Our preliminary data show that the trapping tends to occur in regions where the gradients in intensity are greatest such as at the edge of the shadow of the aperture that is used to define the beam. This is not surprising since most of the light trapping arguments utilize gradients in the fields to initiate the process. We currently are attempting to observe the dynamics of this trapping and also to correlate the trapping dynamics with the growth of the Stoke and anti-Stokes waves. This should considerably clarify the whole process.

It still remains to be seen if this will explain the "wrong cone angles" present in the anti-Stokes emissions, as well as some of the other problems that have not yet been explained. However, it does appear that the beam divergence of the Stokes wave and the angular breadth of the anti-Stokes cones may be due to the diffraction spreading from these guided waves.

SUMMARY OF THEORETICAL WORK

Theoretical studies are underway to understand the observed thresholds for nearly-forward-directed stimulated Raman scattering and the angles and spectra of the various components of this forward stimulated emission. At the present, there is no clear idea as to which underlying mechanisms most affect threshold and emission characteristics, so our studies thus far have comprised a series of exploratory examinations of all mechanisms which we felt could not be ruled out on the basis of the experimental evidence to date. Since at this stage it would be of little use to give the details of any of these studies, we will only indicate here the various avenues which we are exploring.

If the incident laser beam were greatly compressed or "trapped" into small filaments by some non-linear process in the Raman medium, then many puzzling features of Raman scattering could be understood. That electrostriction could cause such "self-trapping" of strong light beams is well known. However, an accurate estimate of the amount of trapping has not been made, and we are looking for ways to make it. That other mechanisms, such as the electronic contribution to the third order non-linear polarization, could produce such "self-trapping" is also well known, but quantitative estimates of the amount are again lacking. The non-linear equations are actually quite different for trapping by electrostriction or by the non-linear electronic susceptibility, and we have studied some effects of these differences. For a given magnitude of a dimensionless coupling coefficient, the latter effect should be much more effective in producing trapping than the former because of the greater localization in space of the electronic interaction.

If the ordinary Raman scattering peak cross-section were much larger in the forward direction of scattering than in other directions, then this would explain many puzzling features. Such an anomalously large forward scattering cross-section would occur in plasmas because of the long range collective excitations (electron and ion plasmas). If a long range collective excitation based on molecular vibration should exist in liquids, we would similarly expect anomalously large forward Raman scattering. We are exploring the possible existence of such a state.

It is well known that the parametric coupling of the Raman waves to acoustic or other such matter or light waves could alter the apparent gain upward from the normal gain expected for stimulated Raman scattering. We have found over half a dozen different waves which might partake in the increase in Raman gain and have developed the formal framework to study them.

We have also tried to formulate theories which might describe the effects on Raman scattering of local heating, shock waves, and field gradients normally present in an incident beam. In every instance, we look for experiments which could shed some light on the effect in question and try to order such possible experiments on the basis of their difficulty and potential significance. Hopefully, a narrowing of the field of possible pertinent effects will enable us to concentrate more theoretical effort on fewer topics in the future.

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Comparison of Observed and Predicted Stimulated Raman Scattering Conversion Efficiencies*

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We have measured the absolute Raman scattering cross section for the 1345-cm^{-1} shifted Stokes line in nitrobenzene using a ruby laser. We have also measured the efficiency of conversion of radiation from a giant pulse ruby laser into stimulated Raman scattered radiation for this line. We have found that the average intensity needed to produce a given conversion is one or two orders of magnitude less than that predicted by the theory of Hellwarth using our measured cross section. We discuss some possible explanations for this disagreement.

INTRODUCTION

USING the absolute Raman scattering cross sections measured by McClung and Weiner[1] and approximate values for the pump laser power density needed to obtained stimulated Raman scattering (SRS) in the initial SRS experiments,[2] Hellwarth[3] showed rough agreement between observed conversion efficiency and that predicted by his theory. Because the prediction of conversion efficiencies from the Raman cross sections provides a crucial test of the present theories of SRS we decided to make more accurate conversion efficiency measurements. These were made with the Raman material outside the laser cavity, not inside as in the initial SRS experiments.[2] They were found to disagree with the predicted conversion efficiencies by one or two orders of magnitude. In this paper we describe our measurement of conversion efficiencies and our remeasurement of the absolute Raman cross section in nitrobenzene. We also show how the conversion efficiency may be predicted from the cross section under various assumptions. Finally, we discuss some proposed explanations for the discrepancy between predicted and observed conversion efficiencies.

THE RAMAN SCATTERING CROSS SECTION

After initial conversion efficiency measurements revealed a discrepancy it was thought that the absolute Raman cross sections might be in error. Hence it was decided to measure the cross section of the 1345-cm^{-1} shifted nitrobenzene Raman line. This line was chosen as it is much wider than the laser line, and also its shape was well known from our previous experiments so that only the total cross section need be measured.

The experimental arrangement for this measurement is shown in Fig. 1. The beam from a ruby laser was directed through a condensing lens onto a rectangular cell containing nitrobenzene. Its polarization was perpendicular to the plane of the laser beam and the direction of observation of scattering. Its energy was about 1 J and average power about 10 kW. The Raman

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[1] F. J. McClung and D. Weiner, J. Opt. Soc. Am. **54**, 641 (1964).
[2] G. Eckhardt, R. W. Hellwarth, F. J. McClung, S. E. Schwarz, D. Weiner, and E. J. Woodbury, Phys. Rev. Letters **9**, 455 (1962).
[3] R. W. Hellwarth, Appl. Opt. **2**, 847 (1963).

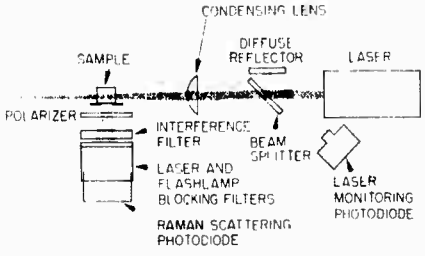

FIG. 1. Schematic of experimental arrangement for measuring the Raman cross section.

scattered energy was detected by a $\frac{1}{4}$-cm² silicon photodiode after passing through a polarizer, an interference filter of ~100-Å bandwidth peaked near 7660 Å, and various Corning glass filters that absorbed laser and flashlamp radiation. To obtain an absolute cross section, the above Raman scattering was compared with the scattering from a MgO surface (set at 45° with respect to the laser and observation direction) that replaced the sample. For this measurement the sample-to-photodiode distance was increased by a factor of 20 and the various filters and polarizers were replaced by four Wratten ND1 filters. Electronic circuit parameters were left unaltered. The integrated signal from the scattering detector was displayed on one trace of a dual beam oscilloscope. The integrated signal from the laser monitor photodiode was displayed on the other trace so that the scattering readings could be normalized to a given laser input power.

The filters and polarizers were calibrated at their appropriate wavelengths (6943 Å or 7660 Å) with a Bausch & Lomb 2-m spectrograph that had been converted into a spectrometer. The interference filter was aligned by an autocollimation technique. The spectral response of the scattering detecting silicon photodiode was measured by comparing its response to the response of a thermopile for light transmitted by various interference filters.

Various relative measurements were performed to reduce the possibility of systematic errors in the experiment or its interpretation. The addition of a filter that transmitted only ruby laser radiation completely eliminated the output of the scattering detector in the Raman scattering measurement. This showed that scattered ruby radiation did not contribute to our signal. The substitution of a broad band (200 Å) interference filter for the narrow band (100 Å) filter indicated that part of the original scattering signal was due to Raman lines other than the 7660-Å line. A knowledge of the frequency response of the two filters and a crude knowledge of the Raman spectra enabled us to calculate accurately that 81% of the original signal was due to the 7660-Å line. The power density on the Raman sample was varied while keeping the laser power fixed by moving the condensing lens with respect to the sample. For changes of power density of more than a factor of 100, no changes in the amount of scattering could be observed. Finally, the Raman scattering was observed at an angle of about 12° from the laser beam in the backward direction. Within error, the scattering was isotropic.

The Raman scattering cross section was calculated from the following formula:

$$K=\frac{D_1M_2R_1^2A_2B_2\cos\theta n^2}{D_2M_1R_2^2A_1B_1\pi L}. \qquad (1)$$

Here subscript 1 refers to Raman scattering measurements at 7660 Å and subscript 2 to the measurements of scattering of 6943-Å radiation from the MgO. K is the total scattering (integrated over wavelength) per unit length per unit solid angle in the medium. D is the voltage from the scattering detector, and M is the voltage from the laser monitor. R is the distance from the sample to the photodiode after it has been corrected for the refractive index of intervening materials. A is the attenuation of the filters and polarizers used at the appropriate wavelength. B is the current per unit power at the appropriate wavelength. $\pi^{-1}\cos\theta$ is the factor from Lambert's law (which has been well verified by Giordmaine[4] for laser intensities) for scattering from the MgO surface whose normal is at an angle θ with respect to the direction of observation. n^2 is the square of the nitrobenzene refractive index used to correct external to internal solid angles. Finally, L is the length of the scattering cell illuminated by the laser beam that is seen from the photodiode.

The result for the wavelength integrated cross section was 5.0×10^{-8}/cm-sr. (This corresponds, for isotropic scattering, to a cross section of 10^{-28} cm² per molecule.) The shape of the 7660-Å Raman line has been measured, during our previous absolute cross-section measurements[1] (although the precise shape was not published). From this we determined that a rectangular line of width 6.0±0.5 Å (measured outside the medium) whose height was the peak height of the Raman line, had an integrated intensity equal to that of the Raman line.[5] The width inside the medium would be 3.9 Å. Hence the peak differential scattering cross section is $K_{\max}=5.0\times10^{-8}/3.9\times10^{-8}=1.3$/cm² sr. The estimated error is ±30%. This result agrees within error with our previous measurement[1] of 2.3±1.1 cm⁻² sr⁻¹.

OBSERVED CONVERSION EFFICIENCY

The experimental arrangement for measurement of the conversion of giant pulse radiation (6943 Å) to first Stokes radiation in nitrobenzene (7660 Å) is shown in Fig. 2. Collimated light from a ~10-MW giant pulse laser is made to pass through a 1.45-mm-diameter aperture

[4] J. A. Giordmaine (private communication).

[5] Due to a small numerical error, the linewidth should be 9 cm⁻¹, not 11 cm⁻¹ as given in Table I of Ref. 1 for this Raman line. The other entries in the table are correct.

into a cell containing nitrobenzene. The aperture is placed in the region of maximum intensity of the beam, this region being determined by burning polaroid film with the much attenuated laser. The laser light and the SRS that has been generated are allowed to fall on a a MgO block. The light scattered from this block is detected by two separate photodetectors.

The photodetector for the 6943-Å light was a Korad "bomb" photodiode. Its calibration was checked against two different kinds of calorimeters and all agreed to within about 10%. A silicon photodiode was used to detect the 7660-Å light. It was calibrated against the "bomb" diode at 6943 Å, and its sensitivity at 7660 Å was determined as noted above. A 7660-Å norrow-band interference filter plus several Corning glass filters insured that only first Stokes radiation entered the silicon photodiode. As a check, the addition of a 6943-Å narrow-band filter to these filters was shown to reduce the first Stokes signal to zero. All filters were calibrated in the way noted above.

The main results of this experiment are shown in Fig. 3. Here the average power density over the aperture is plotted against the fractional conversion of 6943-Å power to first Stokes power. The cells were carefully tilted so that their windows were 1° from being aligned perpendicular to the laser beam.

Several other points, not shown in the figure, were taken with the 10-cm cell for tilt angles between 1° and 6°. These points for a given angle setting fell on curves roughly parallel to those shown. These curves were in all cases between those shown in the figure and closer to the 10-cm cell curve than the 15-cm cell curve. There appeared to be no correlation between angle and conversion efficiency. This is probably because the diffuse scattering of the window that is responsible for feedback depends more on the quality of the part of the window that the laser happens to strike than upon the orientation of the window. When a glass flat was placed after the tilted 10-cm cell and aligned optically parallel to the 45% reflector at the end of the laser, the conversion efficiency (measured in the 20-MW/cm² region) approximated that of the tilted 10-cm cell.

For completeness, we note the following result for total conversion efficiency when the aperture is removed from in front of a tilted 10-cm cell. Here a laser beam of about 10^{-3} rad beam divergence with a radial power distribution roughly approximated by $\exp[-(\text{radius}/0.3\text{ cm})^2]$ is passed through a 2.3 power beam diameter reducing telescope. About 10% of a 15-MW laser beam was converted to first Stokes power.

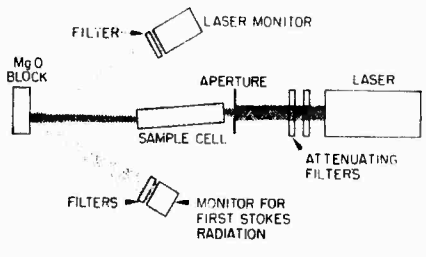

FIG. 2. Schematic of experimental arrangement for measuring conversion efficiencies.

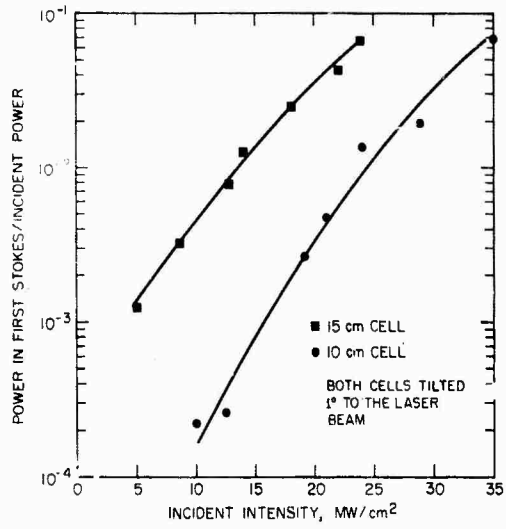

FIG. 3. Efficiency of conversion to first Stokes radiation in nitrobenzene.

PREDICTED CONVERSION EFFICIENCY

If a plane wave of initial intensity N propagates a distance l through a medium of gain per unit length g_β and effective loss L, its intensity at the end of this distance is $Ne^{g_\beta l - L}$. According to Hellwarth[3] the gain per cm for SRS is given by

$$g_\beta = I_a[S_{\alpha\beta}(N_1 - N_2)]\lambda_\beta^4/C_\beta, \qquad (2)$$

where I_a is the number of photons/cm²-sec within the wavelength spread of the Raman line that is incident on the material at the pump frequency, λ_β is the wavelength of light in the medium at the Raman-shifted frequency, C_β is the speed of light in the medium at this frequency, and $S_{\alpha\beta}(N_1 - N_2)$ is the peak scattering cross section per unit volume per steradian per unit wavelength in the medium. This implies a gain of 1.4×10^{-3} cm^{-1} for a 1-MW/cm² pump and our 1.3 cm^{-2} cross section. We note that the units of cross section (cm²) per unit volume (cm^{-3}) per unit wavelength (cm^{-1}) are cm^{-2}.

The initial intensity (noise) to be amplified by the Raman material is of order 10^{-5} to 10^{-6} W/cm². The lower estimate represents the Raman scattering into a 1/10-Å bandwidth (at 7660 Å) and 10^{-5} sr solid angle by a 10 MW/cm² ruby laser beam striking a 10-cm cell of nitrobenzene. The higher estimate represents the quantum-mechanical noise associated with zero-point vibrations in the electromagnetic field.[6]

[6] W. G. Wagner (private communication).

We have found that the backscatter from our cell a few degrees from the direction of incidence of the laser beam (and from the direction of specular reflection) is about 1% of the scattering from a MgO surface. This was also true of a thin pice of glass. This fact enables us to estimate effective reflectivity R_2 below, which we take as just 1% of the Lambert's law scattering at normal incidence into the solid angle of acceptance of the laser.

We may estimate the gain/cm, g_β, using the formula

$$fP/N=(R_1R_2\exp[2lg_\beta])^m/R_2. \quad (3)$$

Here f is the fraction of the incident power converted, P is the incident power/unit area, and N the initial noise power/unit area, R_1 and R_2 are the effective reflectivities of the cell windows, l is the cell length, and m the number of (double) passes the wave makes during amplification. These passes are assumed to start from noise at reflector 2 and travel toward reflector 1, the output reflector of the GPL.

At 25 MW/cm², about 1% of the initial energy is converted in a 10-cm cell. Under the assumptions of low noise ($N=10^{-6}$ W/cm²) and small solid angle of acceptance ($\sim10^{-6}$ sr), which imply low reflectivity ($R_1=0.5$, $R_2=3\times10^{-9}$) we find the gain per cm $g_\beta=1.0+(0.3/m)$. For the case of higher noise ($N=10^{-5}$ W/cm²) and a larger solid angle (10^{-5} sr implying $R_2\sim3\times10^{-8}$) we find $g_\beta=0.9+(0.3/m)$. (Here m is less than 5 for our geometry and GPL pulse length.) For the case of a sing. one-way pass we find $g_\beta=2.4$ cm^{-1}. From our cross section we expect $g_\beta=0.035$ cm^{-1} at 25 MW/cm². Hence the gain deduced from the conversion efficiency is one or two orders of magnitude too high and is rather insensitive to the specific assumptions used for its calculation.

We can also show from our conversion efficiency curves that dg_β/dP is not a constant as is predicted by Eq. (2) and that at lower powers it is much higher. To compute the gain from the slope of the conversion efficiency curve we derive from formula (3) the following expression for the ratio of power conversion at input power/area P_1 and P_2:

$$f_1P_1/f_2P_2=\exp[2(dg_\beta/dP)(P_1-P_2)lm]. \quad (4)$$

Assuming dg_β/dP constant, this implies $g_\beta\sim0.3$ cm^{-1} for $m=1$ and $g_\beta\sim0.06$ cm^{-1} for $m=5$, both for $P\sim25$ MW/cm². Hence, the gain computed this way is 4 to 15 times smaller than the average gain needed to achieve the observed conversion starting from noise.

We indeed observe an onset of the increasing slope in our Stokes power vs GPL power curves as we decrease our laser power. Bret,[7] however, has actually measured Stokes power to a level about 10^5 times lower than we did, and finds a very sharp increase in slope at low GPL power. His measurements for a 20-cm cell of nitrobenzene agree very well with ours for a 10-cm cell where our data overlap. That our 10-cm cell and his 20-cm cell give the same results (within experimental error) is not surprising because there were slight differences in the experimental arrangements. (Quantitatively, he observes 10^3 W/cm² of Stokes output for 10 MW/cm² of GPL input. However, he sees 1 W/cm² of Stokes at 5 MW/cm² GPL and 10^{-2} W/cm² of Stokes at 3.5 MW/cm² GPL.)

DISCUSSION

There are many possible ways we might attempt to explain the above discrepancy. One class of explanations involves the incompleteness of the theory. For example, the inclusion of a four photon process,[8] of nonresonant coupling,[6] of the miltimode character of the GPL pump in the theory might help to resolve the present disagreement. Another class of explanations assumes errors in the estimation of parameters used in finding g_β. One of these is an incorrect estimation of the intensity of the GPL pump caused by nonuniform energy distribution in the beam, perhaps due to optical trapping.[9] Another is an incorrect estimation of the reflectivities. This might be due to light reflection from sound waves produced in stimulated Brillouin scattering.[10] We discuss these possible explanations in more or less detail below. However, it is clear that more detailed theoretical work and additional controlled experiments are required to determine with confidence the correct explanation.

The four photon process involves the conversion of 2 pump photons into a Stokes photon and an anti-Stokes photon. The gain for this process should be of the order of magnitude of the ordinary SRS gain near the index matching directions but less in other directions. Hence, the gain in the direction of the laser beam is smaller so that this process, while it may contribute to the discrepancy, is probably not primarily responsible.

The multimode character of our GPL may have an important effect on conversion efficiency. Hellwarth's theory is applicable only to a single-frequency laser pump, and we know that even a small multimode character in the laser can have large effects on various aspects of SRS. For example, spreads of less than one cm^{-1} in the laser frequency cause spreads of more than 100 cm^{-1} in the anti-Stokes frequency, and considerable spreads in anti-Stokes emission angle.[11,12] Although no quantitative measurements of single mode conversion efficiencies have yet been made, qualitative observations

[7] G. Bret, Z. Angew. Math. Phys. **16**, 33 (1965).

[8] S. Yatsiv (Private communication).

[9] R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Letters **13**, 479 (1964).

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[11] B. P. Stoicheff, Phys. Letters **7**, 186 (1963).

[12] R. W. Hellwarth, F. J. McClung, W. G. Wagner, and D. Weiner, Z. Angew. Math. Phys. **16**, 27 (1965).

suggest that single mode behavior by itself is not sufficient to account for the large discrepancy.

We now consider evidence consistent with the assumption of nonuniform energy distribution in the beam. We emphasize again that none of this evidence is conclusive and that further experimentation is needed.

There is direct experimental evidence for some nonuniformity of the GPL beam. Magnified near-field photographs show a granular structure with some regions at least twice as intense as the average intensity over an area the size of the aperture. This does not preclude greater intensity variations on a finer level of resolution in space and time.

There is also theoretical reason to believe that high-intensity light beams tend to become self-trapped and hence increase the peak intensity of the beam. Chiao *et al.*[9] estimate that these effects take place at powers well within those reached in our experiments. They predict that the self-trapping threshold depends inversely on the nonlinearity of the refractive index. Since nitrobenzene has stronger nonlinear behavior than benzene, this would explain why their SRS thresholds are about the same, although the peak absolute Raman cross section in nitrobenzene is smaller than in benzene.

The addition of a carefully aligned specular reflector after a cell that gave $\sim 0.3\%$ conversion at ~ 20 MW/cm^2 before the reflector was added gave only $\sim 3\%$ conversion at this intensity afterward. Since for a uniform beam the conversion should be proportional to R_2^{m-1} and since R_2 was increased from $\sim 10^{-8}$ to $\sim 10^{-2}$, one would expect SRS conversion to approach unity. However, we see that we can construct examples of nonuniform intensity distribution that have the correct average intensity and give the correct conversion using the measured cross section. For example, any distribution in which 0.3% of the energy was in regions of at least 30 times the average intensity and in which 3% of the energy was in regions of at least 20 times the average intensity would give this result. This result was computed assuming all the SRS came from a region of near unity conversion and that there were three effective double passes. The latter, not unreasonable, assumption implies a 3:2 ratio of g_β in the two regions at 20 MW/cm^2. For the same distribution of energy, this implies $\sim 3\%$ conversion at ~ 30 MW/cm^2 for the case $R_2 \sim 3\times 10^{-8}$ as was observed.

Dennis and Tannenwald[13] have performed an experiment in which the effects of nonuniformity along the GPL beam should average out to the uniform illumination case. They focus a 2-MW GPL with a cylindrical lens into a line focus in nitrobenzene and observe SRS along this line when they place high-reflectivity mirrors perpendicular to it. They estimate a gain which corresponds to ~ 0.02 cm^{-1} at 25 MW/cm^2. This gain should only be slightly less than the gain in the direction of the GPL beam.[6] Hence their gain agrees within error with our theoretical estimate. However, this experiment can also be used to support a different hypothesis as noted below.

The present experiments, the initial SRS experiments,[2] and the experiments of Dennis and Tannenwald[13] indicate that when higher reflectivity end reflectors are used, better agreement between observed and predicted conversion efficiencies are obtained. This is consistent with the hypothesis that some physical effect is causing the effective reflectivity in our low-reflectivity experiments to be much higher than we had originally supposed. If we accept the theoretical values for g_β and the experimentally measured threshold for SRS we need an effective reflectivity around the Raman material of about 80% and oriented so that specular reflection will occur directly back to the laser. One possible way to obtain such a reflectivity is by stimulated Brillouin scattering.[10] The intense acoustic waves generated by the stimulated Brillouin scattering form a three-dimensional grating which scatters Stokes and anti-Stokes energy in addition to the laser energy. This presupposes that there is indeed stimulated Brillouin scattering occurring in materials that show abnormally low SRS thresholds. Such an assumption can be easily checked experimentally. It is, however, necessary that such checks be performed at or near the SRS thresholds. If no such stimulated Brillouin scattering exists, then the existence of other acoustic waves which could be produced may also serve the same purpose. Such checks are currently being made in this laboratory. If stimulated Brillouin scattering is taking place and also aiding in the regeneration of the Stokes radiation, then the experiment should show that the backscattered Stokes radiation is red shifted by the Brillouin frequency for the material.

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MODE-STRUCTURE INDEPENDENCE OF STIMULATED RAMAN-SCATTERING CONVERSION EFFICIENCIES*

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The power density needed to convert a given amount of laser energy to stimulated Raman-scattered radiation has been found to be about an order of magnitude less in nitrobenzene than theoretically expected for the case of a Raman cell external to the laser cavity.[1] It has been suggested[1,2] that the multimode character of the laser pump might be responsible for this disagreement. Bloembergen and Shen[2] have given a semiquantitative estimate of the enhancement of the Raman gain. It indicates that with a typical multimode laser a Raman gain ~4 to 8 times greater than that for a single-mode laser should be produced. It is the purpose of this communication to show that the Raman gain in nitrobenzene for a single-mode laser pump is precisely that for a multimode laser pump, and to suggest other reasons for the above-mentioned disagreement.

In this experiment, the collimated beam from our giant pulse laser was directed onto a 1.45-mm aperture 70 cm from the laser, and the portion of the beam transmitted by the aperture passed through a 10-cm cell of nitrobenzene onto a MgO diffuse reflector placed 50 cm after the cell. The power of the laser and the first Stokes line was monitored by suitably filtered fast photodetectors that sampled the diffuse reflection from the MgO. The cell was tilted at ~3° with respect to the laser beam. This arrangement is like that described in more detail by Weiner, Schwarz, and McClung,[1] except that the aperture and cell are now further from the laser and closer to the MgO. Also, another beam splitter to allow a measurement of the far-field pattern has been added.

Our laser has been mode-selected to give single-transverse and longitudinal mode behavior. The details of the mode-selection techniques will be given elsewhere.[3] The transverse mode structure was determined with the aid of a 1-m focal-length camera. The longitudinal mode structure was determined with the aid of a 2-cm spaced Fabry-Perot etalon with $\lambda/80$ flat plates of ~1% transmission. When completely mode-selected, the laser produced 2 MW of power in a beam whose divergence equaled the diffraction limit corresponding to the laser-beam diameter. When not mode-selected, the laser output was ~10 MW, the beam divergence $\sim 1.5\times 10^{-3}$ rad, and the spectral width $\sim\frac{1}{2}$ cm^{-1}. The pulse length was ~30 nsec for both cases. The results of the conversion-efficiency measurements for mode-selected and non-mode-selected lasers are shown in Table I. The experimental arrangement is the same for both cases. The relative error for the power measurements is ~5%. Measurements at other power densities for our mode-selected laser gave a conversion-efficiency curve which agreed very well with our previous curve for a non-mode-selected case.[1] The data of Table I indicate strongly that the anomalously high gain is not caused by the laser mode structure.

The theoretical gain in nitrobenzene at 20 MW/cm is 0.028 cm^{-1}. This gain is computed using a formula of Hellwarth[4] and a recently measured peak Raman-scattering cross section of 1.3 ± 0.4 cm^{-2}.[5] This cross section agrees within the expected error with our earlier and less accurate measurement of 2.3 ± 1.2 cm^{-1}. It also agrees with the recent measurement of Damen, Leite, and Porto[6] in the fol-

Table I. Conversion efficiency for mode-selected and non-mode-selected pumping of nitrobenzene.

Description of mode structure	Incident-peak power density (MW/cm^2)	Power converted to first Stokes radiation (%)
Two TEM_{00} modes of ~4:1 intensity ratio separated by ~800 Mc/sec	19	0.6
~100 transverse modes ~10-20 longitudinal modes	19.5	0.6

lowing sense. If we scale our data for benzene cross sections by the ratio of the nitrobenzene cross sections (1.3/2.3) and then compute the cross section/molecule at 6328 Å, we find a value of $0.7\pm0.2\times10^{-28}$ cm^2. Here we assume a λ^4 wavelength dependence and use the observed angular dependence for Raman scattering. This value agrees (within the stated error) with their[6] value of $0.56\pm0.1\times10^{-28}$ cm^2.

To estimate the Raman gain at 20 MW/cm^2 from our conversion-efficiency data, we assume noise is amplified during a double pass (through the cell to the laser reflectors and back through the cell) to the observed level of 10^5 W/cm^2. The dominant contribution to the noise which initiates the generation of the Raman-shifted radiation comes from the quantum-mechanical zero-point vibrations of the electromagnetic field. In a spectral interval of $d\lambda$, and within a cone of opening angle θ, centered on the axis of the laser beam, there is a zero-point power flux[7] given by $(\pi\theta^2\hbar\omega/n\lambda^3)(d\lambda/\lambda)$. Our measurements indicate that the bulk of the Stokes radiation emerges within an angle, θ, which is not greater than 0.017 rad, and that the fractional wavelength spread, $d\lambda/\lambda$, is of order 3×10^{-4}, and thus the starting power flux is estimated to be $\leq1.2\times10^{-2}$ W/cm^2. Then 10^5 $W/cm^2 = 1.2\times10^{-2}$ W/cm^2 $\times\exp(g\times20\text{ cm})$, so that the gain per cm $g = 0.80$ cm^{-1}. The gain is only slightly lower if calculated for multiple double passes where the feedback is supplied by diffuse reflection from the cell window and the MgO reflector, since the solid angle of acceptance imposed by our geometry is so small. Thus the observed gain is at least 25 times greater than that calculated from the cross sections.

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[7]It is assumed that the Raman radiation is strongly polarized, in accord with the experiments. The wavelength to be used in the formula is that of the Stokes radiation, inside the Raman cell, and n is the linear index of refraction at the Stokes frequency ω.

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The problems associated with the production of diffraction-limited Raman based laser-like devices have been under investigation. The most persistent effort has been directed at understanding several of the rather serious deviations between theory and experiment. The most noteworthy problem treated here is the anomalous gain of Stimulated Raman Scattering (SRS). We have analyzed several approaches, theoretically and experimentally, with some success in only one approach. At present, it appears that the solution is contained in considering light trapping effects as the cause of a highly increased power density in the Raman material. Currently, efforts are continuing to explain the mechanism of the trapping as well as relating these effects to other problems in SRS.

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